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REPORT ON STANDARDIZATION OF MICROCHEMICAL METHODS. CARBON, HYDROGEN, AND NITROGEN

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This year's studies, following the recommendations set forth in last year's report, have been devoted to the determination of carbon and hydrogen and nitrogen by the Kjeldahl and Dumas procedures. The selection was based on the results of a questionnaire, which indicated that in the opinion of a majority of micro analysts these determinations should be studied first.

Two samples, nicotinic acid and benzyl-iso-thiourea hydrochloride, were sent to two groups of collaborators. One group was asked to analyze the samples for carbon and hydrogen; the other was asked to determine nitrogen both by the Kjeldahl and Dumas procedures. These compounds were chosen for this work because both are stable and nonhygroscopic and because they differ considerably in constitution and ease of decomposition.

Although a statement of purity did not accompany the samples, the collaborators were informed that they were relatively pure. They were asked to report all the numerical values obtained for each sample, regardless of whether or not the data appeared to be correct.

The collaborators for carbon and hydrogen were requested to analyze the two samples by their own methods only, since there is no one method in common use today. The collaborators for nitrogen were asked to analyze the two samples by their own Kjeldahl and Dumas methods and by the A.O.A.C. Tentative Microkjeldahl Method.

Questionnaires accompanying the samples asked for details of the apparatus, procedure, and laboratory conditions under which the analyses were conducted. From the information so obtained and the accompanying analytical results, a method will be adopted or devised for each determination, and each will be given rigorous collaborative study before it is proposed as a tenative method. Since so many methods were used in obtaining the data for each determination and consequently the number of variables was so large, the data have been analyzed statistically in an attempt to determine which variations in procedure and apparatus are important. In the statistical comparisons of these data influenced by a large number of variables, the assumption was made that all variables other than the one being evaluated cancelled themselves. The conclusions drawn are based on the available data, and although there is a possibility

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that they may be erroneous because of the assumption on which they are based, they will be used as guides for future studies until more data are available. Since some data were received after the analysis of the data was started, the total number of analyses is in some cases greater than that used in the statistical treatment.

CARBON AND HYDROGEN DETERMINATION

Twenty-five analysts from 19 laboratories reported 111 carbon and hydrogen analyses for sample 1 (nicotinic acid) and 92 analyses for

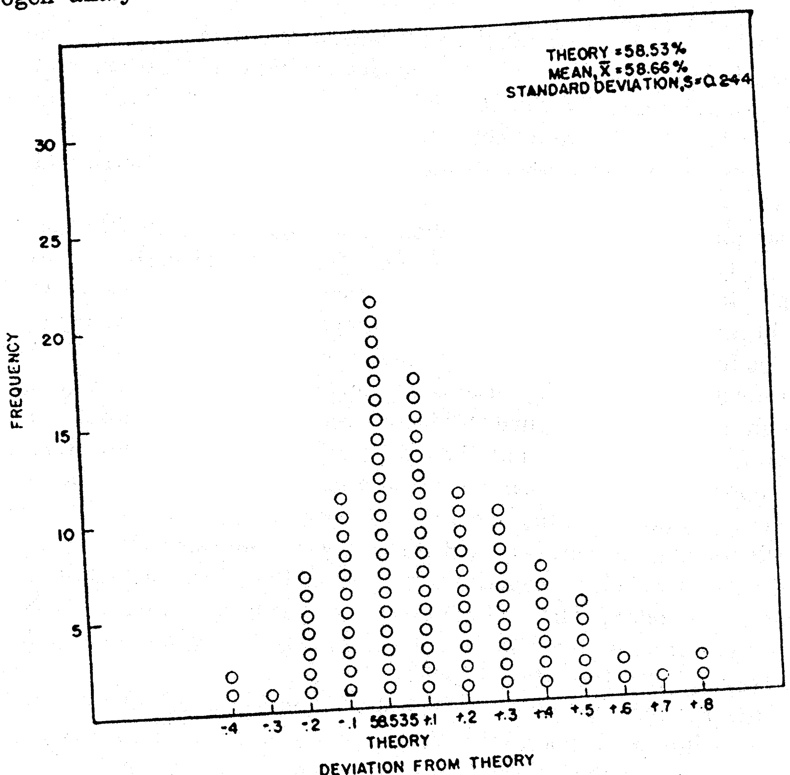


FIG. 1.—Histogram of carbon values for nicotinic acid.

sample 2 (benzyl-iso-thiourea hydrochloride). The number of analyses reported from different laboratories ranged from 2 to 20.

Carbon.—The histogram (Fig. 1) shows the frequency distribution of the carbon values obtained for nicotinic acid. Although all points in the figure are located on the class marks, they show the number of analyses which fell within the class boundaries (class mark $\pm 0.05\%$). Three results are not shown and are not included in the subsequent treatment. The chi square distribution test for measuring goodness of fit of frequency distribution showed that these three values fell outside the representative population of the carbon values for this compound. This test has shown that the values retained in the histogram are a representative sample of a normal population, and therefore they can be treated statistically.

Figure 1 shows that a majority of the values are higher than the theoretical value (58.53%). The mean ($\overline{X}$ or average) of the values is 58.66%, and the standard deviation (S, the variation about a mean) is 0.244, indicating that 67% of the carbon values for this sample should fall within ± 0.244 or 95% within ± 0.488 of the mean value.

Although the mean is 0.13% higher than the theoretical value, the mode (class which contains the largest number of values) falls on theory, indicating that there is a good possibility of finding among the methods one which will give high accuracy as well as good precision. Inspection shows that even though there is a preponderance of values above the theoretical value, the skewness of the histogram toward the theoretical value and the mode indicate that there is a tendency to obtain theoretical values.

All carbon methods followed the same general procedure, which consisted in the catalytic combustion of a weighed sample to carbon dioxide, followed by the absorption and weighing of the combustion product. Although no two methods were identical, each step in any one method was in general similar to the corresponding step in several other methods. Therefore, the carbon values for nicotinic acid were divided into two groups, one representing the results obtained by a certain operation for one step in the determination, and the other a second operation for the same step. These two groups were treated statistically to determine whether or not there was a significant difference in the results obtained by the two operations for the step or condition in question. By this procedure, the following comparisons were made: (1) electrical *vs.* gas sample burner, (2) mechanical *vs.* hand-operated sample burner, (3) semimicro (10–30 mg) *vs.* micro (2–10 mg) sample weight, (4) air-conditioned *vs.* nonair-conditioned laboratories, (5) balance in an air-conditioned balance room *vs.* balance adjacent to the furnace. The only case in which there was a significant difference in the carbon results for nicotinic acid was semimicro *vs.* micro samples. The significance level used throughout this study was 5%, which means that if the experiment was repeated a number of times the differences obtained should be at least as large as the one found in 95% of the cases. The theoretical frequency distribution curves for these two methods are shown in Figure 2. The means ($\overline{X}$) for the two procedures were 58.52 and 58.69, whereas the standard deviations (S) were 0.198 and 0.239, respectively. Since the difference in the two means was significant, and the semimicro mean was closer to the theoretical value, better values can be obtained by the semimicro than the micro method and, similarly, since the standard deviation is less, the precision of the semimicro method is greater.

The data for sample 2, benzyl-iso-thiourea hydrochloride, were treated in the same manner as those for nicotinic acid. Figure 3 shows the histogram of the values obtained for this material. The chi square distribution test showed that the values obtained should all be included in the

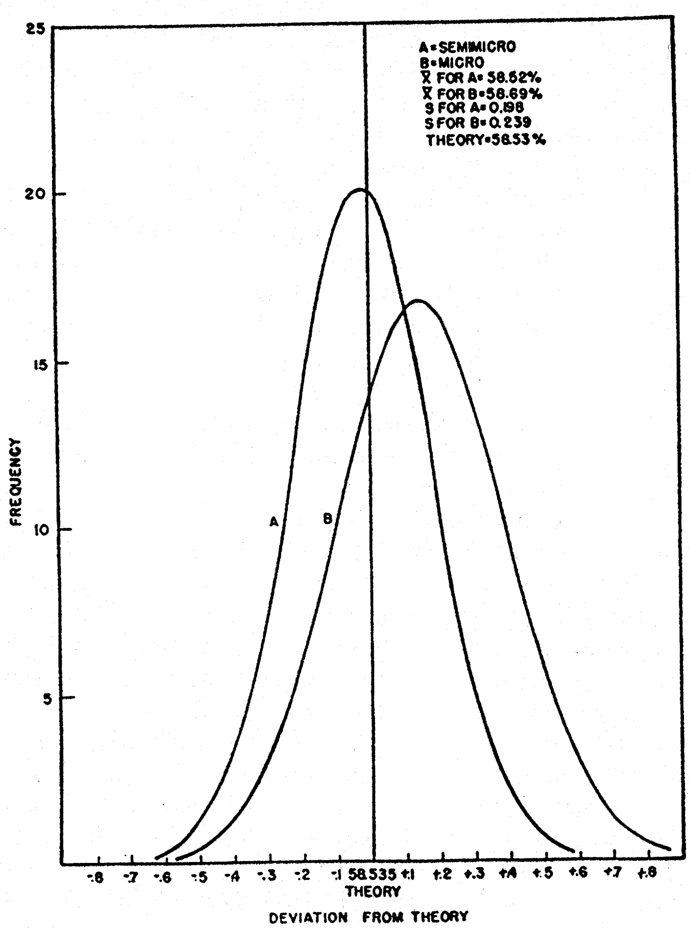


FIG. 2.—Theoretical distribution curves for micro and semimicro carbon values. Nicotinic acid.

statistical treatment. The mean ($\overline{X}$) of the values is 47.51% or 0.11% above theory, with a standard deviation (S) of 0.184. Since the mean is nearer the theoretical value for this sample than that for sample 1 (nicotinic acid), and the standard deviations is less, the methods used gave slightly better carbon values for sample 2. These differences must be due to an inherent difference in the two compounds, since the methods and conditions were the same for the analyses of the two samples. The same comparisons were made for the values for benzyl-iso-thiourea hydrochloride as were made for nicotinic acid. There is again a significant difference between the values for the semimicro *vs.* micro procedures and, as for sample 1, the difference is in favor of the semimicro method. Figure 4 shows the theoretical frequency distribution curves for the two methods. The means are 47.38% and 47.53%, and the standard deviations 0.078 and 0.189 for the semimicro and micro methods, respectively.

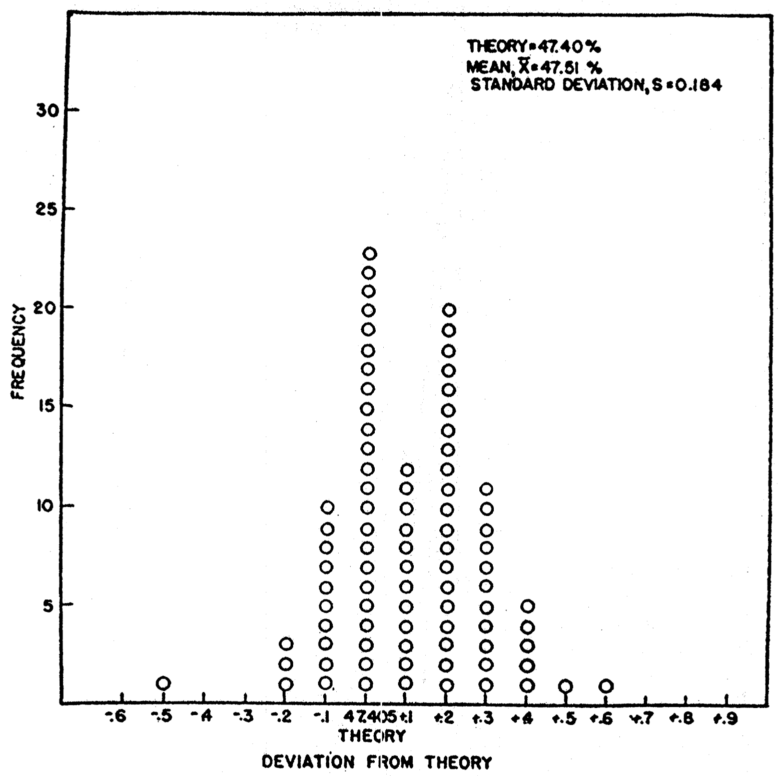


FIG. 3.—Histogram of carbon values for benzyl-iso-thiourea hydrochloride.

The results of the carbon analyses for the two samples are similar, in that the mode in both cases falls on theory, the means are above theory, and the standard deviations for the two are comparable. Semimicro methods were significantly better than micro methods for both samples, as is shown in Figures 2 and 4. None of the other comparisons showed any significant difference which would lead one to recommend the preferential use or adaptation of one procedure over another. Table 1, however, shows slight trends which favor certain operations, and these may serve as guides in planning future studies.

Hydrogen.—The number of hydrogen values received and the number of collaboratores were the same as for carbon, since the two determinations were made simultaneously. Histograms of the hydrogen values for nicotinic acid and benzyl-iso-thiourea hydrochloride are shown in Figures 5 and 6, respectively.

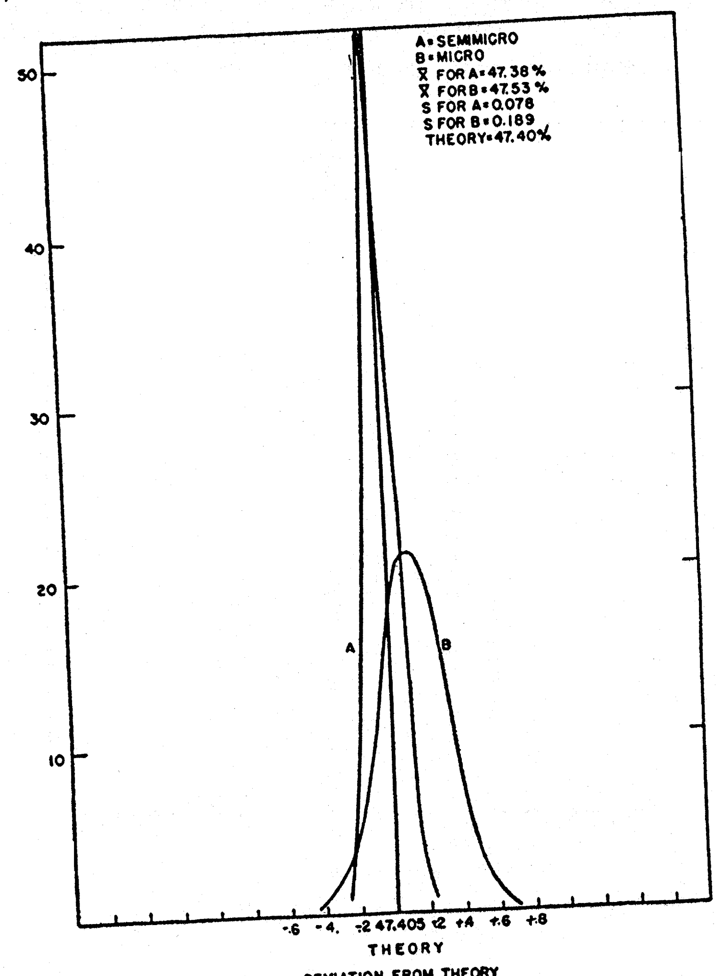


FIG. 4.—Theoretical frequency distribution curves for semimicro and micro carbon values. Benzyl-iso-thiourea hydrochloride.

A marked similarity in the data for the two samples can be seen. The means are 0.10 and 0.06% above the theoretical value, and the two standard deviations are 0.208 and 0.200. In both cases the mode, class with the highest frequency, is 0.10% above theory.

The same operations as listed previously for carbon were compared to determine whether or not they were significantly different. The values

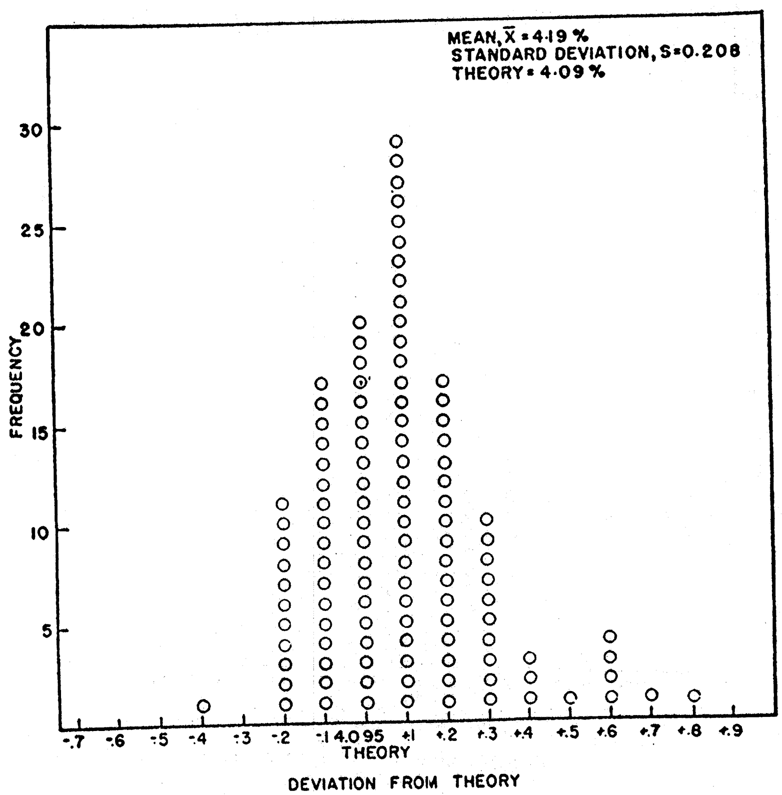


FIG. 5.—Histogram of the hydrogen values for nicotinic acid.

TABLE 1.—*Carbon data obtained for samples 1 and 2 by catalytic combustion methods*

	NICOTINIC ACID (1)			BENZYL-ISO-THIOUREA HYDROCHLORIDE (2)		
	NUMBER OF SAMPLES	$\bar{X}$	S	NUMBER OF SAMPLES	$\bar{X}$	S
		per cent			*per cent*	
Semimicro method	16	58.52	0.198	14	47.38	0.078
Micro method	81	58.69	0.239	76	47.53	0.189
Electric sample burner	50	58.70	0.268	45	47.51	0.148
Gas burner	47	58.62	0.205	44	47.51	0.195
Mechanical burner	41	58.67	0.276	41	47.49	0.143
Hand-operated sample burner	56	58.65	0.209	49	47.53	0.212
Balance in air-conditioned balance room	42	58.72	0.286	34	47.54	0.161
Balance adjacent to furnace	34	58.61	0.221	43	47.47	0.200
Air-conditioned laboratory	59	58.65	0.241	52	47.51	0.176
Non-air-conditioned laboratory	38	58.67	0.179	38	47.52	0.200
Total samples	97	58.66	0.244	90	47.51	0.184
Theoretical values		58.53			47.40	

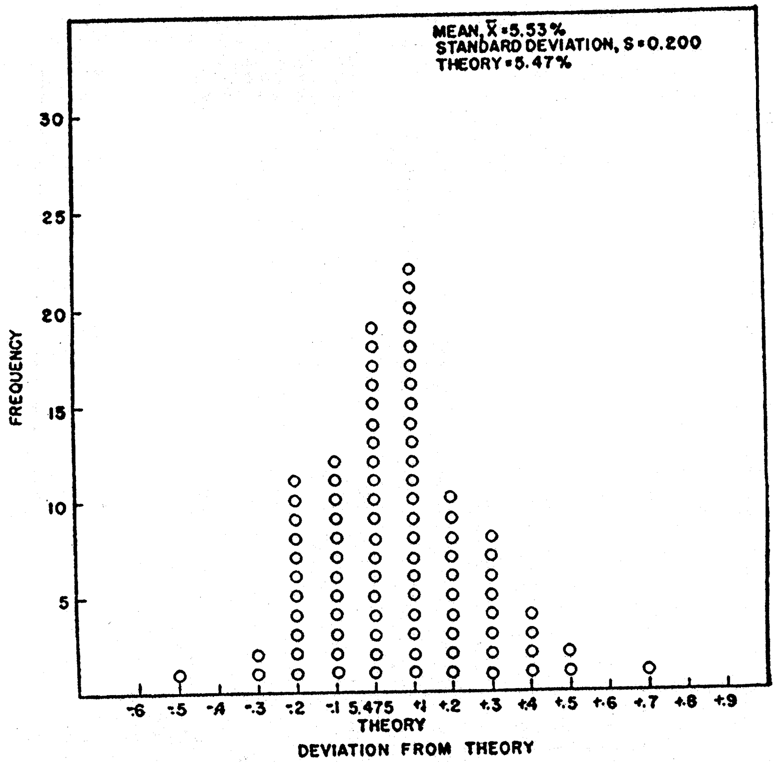


Fig. 6.—Histogram of hydrogen values for benzyl-iso-thiourea hydrochloride.

Table 2.—*Hydrogen data obtained for samples 1 and 2 by catalytic combustion methods*

	Nicotinic acid (1)			Benzyl-iso-thiourea hydrochloride (2)		
	Number of samples	$\bar{X}$	S	Number of samples	$\bar{X}$	S
		per cent			*per cent*	
Semimicro method	18	4.20	0.161	12	5.57	0.109
Micro method	87	4.21	0.214	80	5.53	0.207
Electrical burner	63	4.15	0.175	47	5.51	0.190
Gas burner	52	4.23	0.235	45	5.56	0.205
Mechanical burner	58	4.14	0.195	43	5.48	0.179
Hand-operated burner	57	4.24	0.229	48	5.58	0.210
Balance in air-conditioned balance room	41	4.17	0.176	34	5.50	0.207
Balance adjacent to furnace	44	4.22	0.261	40	5.55	0.219
Air-conditioned	62	4.16	0.178	53	5.49	0.182
Non-air-conditioned laboratory	53	4.23	0.237	39	5.59	0.205
Total samples	115	4.19	0.208	92	5.53	0.200
Theoretical values		4.09			5.47	

shown in Table 2 for mechanically operated sample burners are significantly better than those for hand-operated burners. Figures 7 and 8 show the theoretical frequency distribution curves for mechanical *vs.* hand-operated furnaces for samples 1 and 2, respectively. Only one other comparison, air-conditioning *vs.* non-air-conditioning, proved to be significant. The theoretical frequency distributions in Figure 9 show that

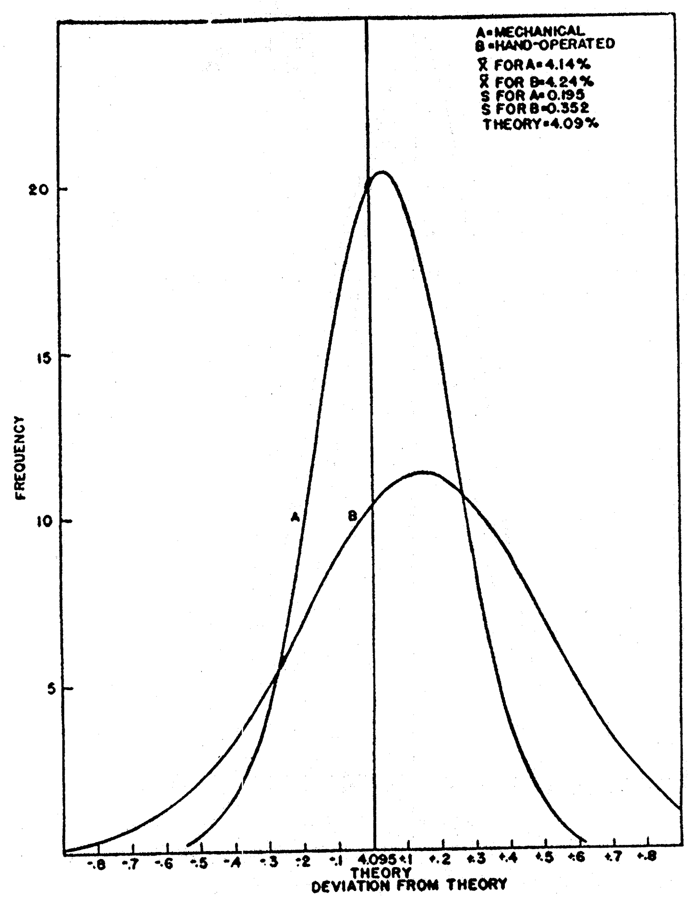


FIG. 7.—Theoretical frequency distribution curves for hydrogen values from mechanical and hand-operated furnaces. Nicotinic acid.

air-conditioned laboratories were superior for the hydrogen analysis of benzyl-iso-thiourea hydrochloride. While the mean is nearer theory and the S value smaller for hydrogen values from air-conditioned laboratories for nicotinic acid, the difference was not significant.

Summary for carbon and hydrogen.—A comparison of the data for hydrogen with those for carbon shows that the means, $\overline{X}$, for hydrogen are slightly closer to theory than those for carbon, that in both analyses the means are above theory, and that standard deviations or precisions for hydrogen and carbon for both samples are similar.

The means for the carbon and hydrogen values obtained in air-conditioned laboratories are closer to the theoretical value than those

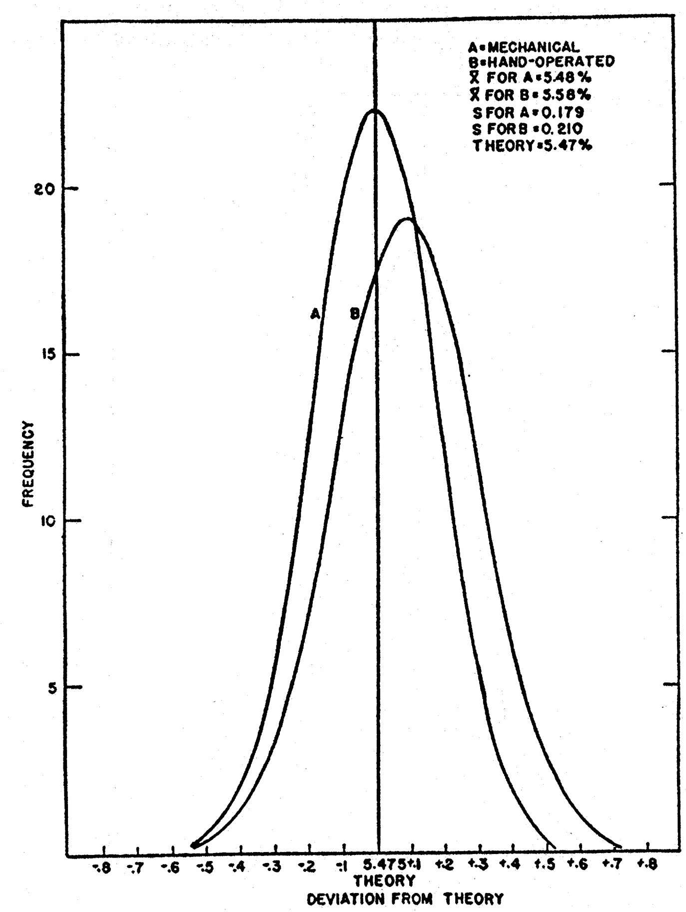


FIG. 8.—Theoretical frequency distribution curves for hydrogen values from mechanical and hand-operated furnaces. Benzyl-iso-thiourea hydrochloride.

made in non-air-conditioned laboratories, but the difference is significant only in the hydrogen values of sample 2. In three of the four determinations, the standard deviation (S) is lower for air-conditioned laboratories.

Comparison of the results obtained by mechanical *vs.* hand-operated furnaces showed that in three of the four determinations, mechanically operated furnaces gave means, $\overline{X}$, closer to the theoretical value and in two cases the difference was significant. In general, the means for electrical burning furnaces are nearer the theoretical value than those for gas burners, and the standard deviations are less, but in no case is the difference significant.

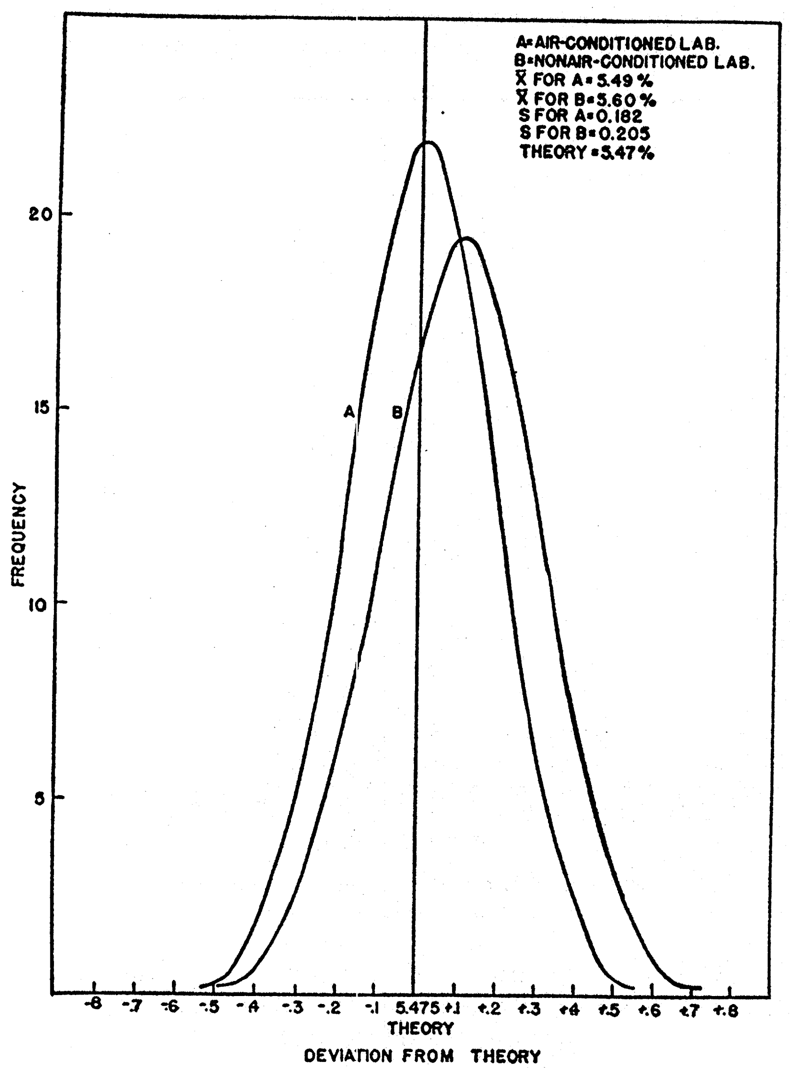


FIG. 9.—Theoretical frequency distribution curves for hydrogen values from air-conditioned and non-air-conditioned laboratories. Benzyl-iso-thiourea hydrochloride.

Perhaps the most significant comparison is that of the values obtained by semimicro and micro procedures for carbon. Here the semimicro procedures are superior, provided that the variables other than size of sample can be neglected. As for the hydrogen values, the standard deviation also favors the semimicro method, and the average values are nearly identical.

Comparison of values obtained when the balance was in an air-conditioned balance room *vs.* those obtained when the balance was adjacent to the furnace shows that the means are nearer the theoretical value and the standard deviations are lower for carbon values when the latter method

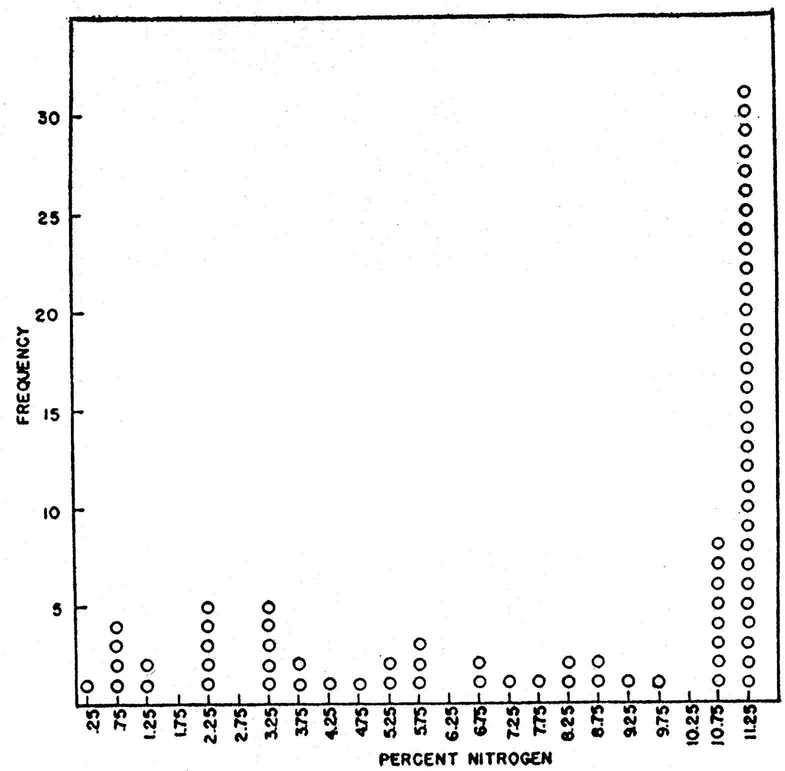


FIG. 10.—Histogram of Kjeldahl nitrogen values for nicotinic acid. Theory = 11.38.

was used, but the reverse is true for the hydrogen values. In no case, however, is the difference between the two procedures statistically significant. More comparisons of the different phases of the analysis, including tube fillings, absorbents, rate of gas flow and the like, are in progress and must be completed before a trial method can be recommended.

KJELDAHL NITROGEN DETERMINATION

The same two compounds used in the carbon and hydrogen studies were sent to a second group of collaborators, who were asked to determine the nitrogen in the two samples by both the Kjeldahl and Dumas procedures.

They were also asked to make the Kjeldahl analyses by their own method and by the A.O.A.C. (tentative) Microkjeldahl Method, a copy of which was enclosed.

One of the reasons for choosing nicotinic acid was that it contains a ring nitrogen, which is difficult to obtain by the Kjeldahl method. Replies to the questionnaire which accompanied the samples indicated that some

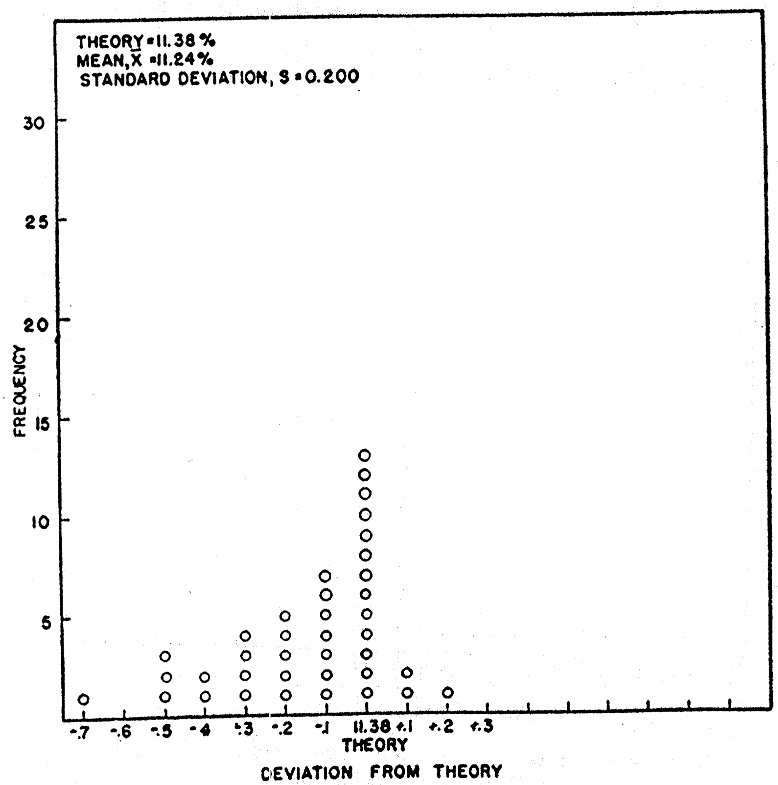


FIG. 11. Histogram of the Kjeldahl nitrogen values for nicotinic acid from the two upper classes in Fig. 10.

micro analysts do not attempt to determine nitrogen in such compounds by the Kjeldahl method but others do it regularly. Therefore, the nitrogen values obtained by the A.O.A.C. method should be a good test of its reliability.

The histogram of the population of the 75 values from twelve collaborators is shown in Figure 10. To present all the data in one histogram, the class intervals had to be in units of 0.5%, since the reported nitrogen values ranged from less than 0.5 to 11.5%.

It is obvious from the histogram that the data as a whole can not be treated statistically but that there is a sharp division of the values, about half being near the theoretical value and the remainder low and erratic.

The collaborators who reported results from 0 to 10% obtained no values higher than 11%, whereas those who reported values about 11% had no values below 10.5%. A histogram (Fig. 11) with class limits of 0.1% was made of the data obtained by those methods which gave one or more values above 11%. Although the mode falls on the theoretical value,

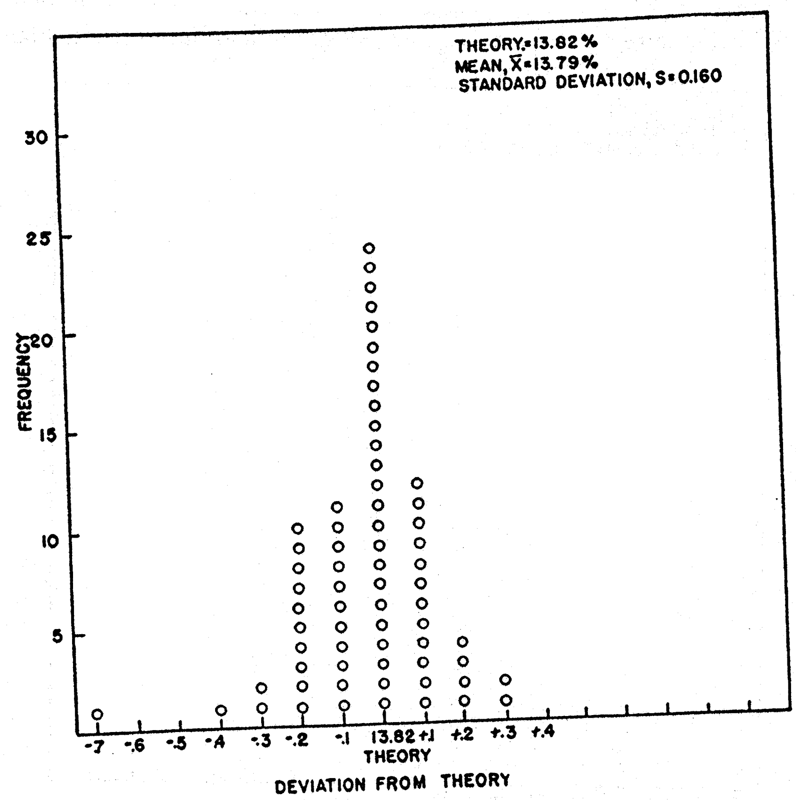


Fig. 12.—Histogram of Kjeldahl nitrogen values for benzyl-iso-thiourea hydrochloride.

the mean is considerably below this value, indicating much skewness in the data. The chi square test showed that the data are not a representative sample of a normal population, and therefore statistical comparisons can not be made. Nevertheless, the means were determined for those values obtained when mercury and mercury plus selenium were used as catalysts, and the mean was closer to the theoretical value when only mercury was used. The many values which are in agreement with the theoretical value indicate that a satisfactory method can be found.

The analysis of benzyl-iso-thiourea hydrochloride (sample 2) for nitrogen by the Kjeldahl method proved to be a much simpler task than determining the nitrogen in nicotinic acid. Figure 12, the histogram of the values, shows that there is a fairly normal distribution of the values around the mode which falls on the theoretical value. The chi square distribution test indicated that the 67 values received are a representative sample and all should be included in the studies. The mean is 13.79, only 0.03% less than theory, and the standard deviation is 0.160.

TABLE 3.—*Nitrogen data obtained for sample 2 by the Kjeldahl method*

	BENZYL-ISO-THIOUREA HYDROCHLORIDE		
	NUMBER OF SAMPLES	$\bar{X}$	S
		per cent	
Semimicro methods	18	13.76	0.212
Micro methods	49	13.81	0.134
Mixed indicator	31	13.77	0.190
Single indicator	36	13.81	0.126
Digestion aid	35	13.80	0.118
No digestion aid	32	13.78	0.195
Electrical digestion	11	13.75	0.224
Gas digestion	56	13.80	0.141
Mercury catalyst	45	13.77	0.167
Mercury plus selenium catalyst	22	13.83	0.134
Parnas-Wargner apparatus	33	13.76	0.167
Other distillation apparatus	34	13.83	0.148
One-half hour digestion	10	13.73	0.228
One hour digestion	57	13.80	0.145
Silver or tin condenser tubes	34	13.75	0.170
Pyrex condenser tubes	33	13.84	0.134
Total samples	67	13.79	0.160
Theoretical values		13.82	

Twenty-three of the values were obtained by the A.O.A.C. procedure, and their mean is 13.795 with a standard deviation of 0.122. The remaining 44 values by various micro and semimicro procedures have a mean of 13.791 with a standard deviation of 0.176. The difference between the means is far from significant, but slightly better precision (smaller S value), was obtained with the A.O.A.C. procedure than with the sum of the other methods.

The data were used to make 8 other comparisons of variations in the Kjeldahl procedure. The comparisons and the mean and standard deviation of each are shown in Table 3. There was no significant difference between the means for any of the comparisons except for silver or tin versus Pyrex glass condenser tubes, the Pyrex glass being favored. The difference between the means for one-half and one hour digestions was nearly significant and in favor of the longer digestion.

The difference between the means for the Parnas-Wagner and other distillation apparatus is considerable, but there is so much duplication of the values obtained by this apparatus and the silver or tin condenser that without further study it is not possible to say whether the lower values are due to the apparatus, the condenser, or both.

Summary for Kjeldahl nitrogen.—The data for nicotinic acid indicate that the A.O.A.C. (tentative) Microkjeldahl method is unsatisfactory for many compounds with ring nitrogen, but it does show that a satisfactory method can probably be established, since four collaborators using four different methods obtained values in agreement with the theoretical values. For the less refractory material, sample 2, the A.O.A.C. method gave values with an excellent mean and with better precision than the sum of the other Kjeldahl methods used. The results indicate that the A.O.A.C. method will apply to compounds with ring nitrogen if the catalyst and digestion time are both increased by amounts yet to be determined.

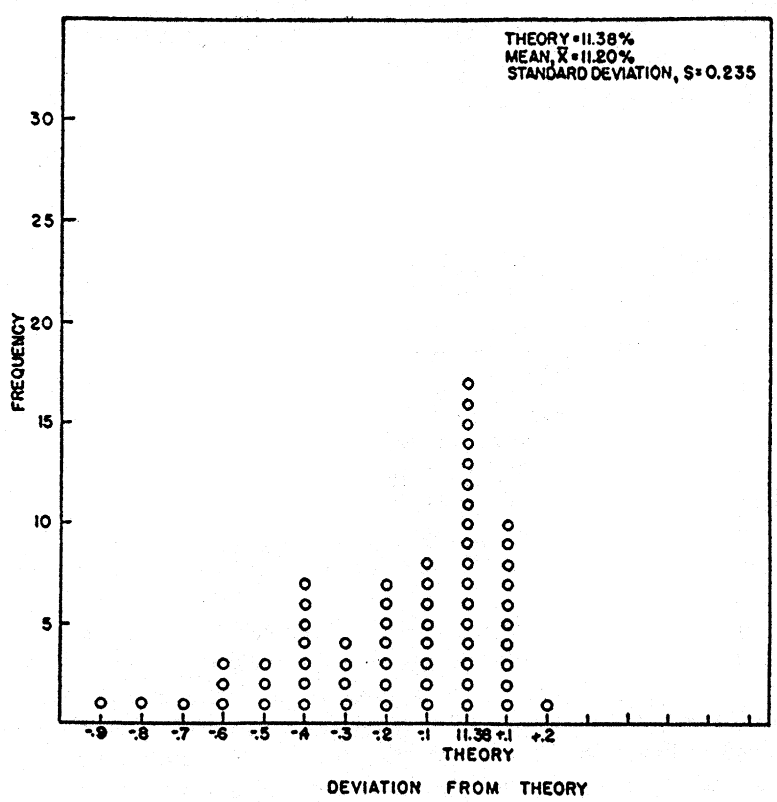


FIG. 13.—Histogram of Dumas nitrogen values for nicotinic acid.

DUMAS NITROGEN DETERMINATION

The same collaborators participated in this determination as in the Kjeldahl studies, and they used the same two samples. Histograms of the values obtained by the 13 collaborators for samples 1 and 2 are shown in Figures 13 and 14, respectively. The values for neither compound were representative samples of normal populations, as shown by the chi square test. Inspection of the histograms shows too many values in the classes

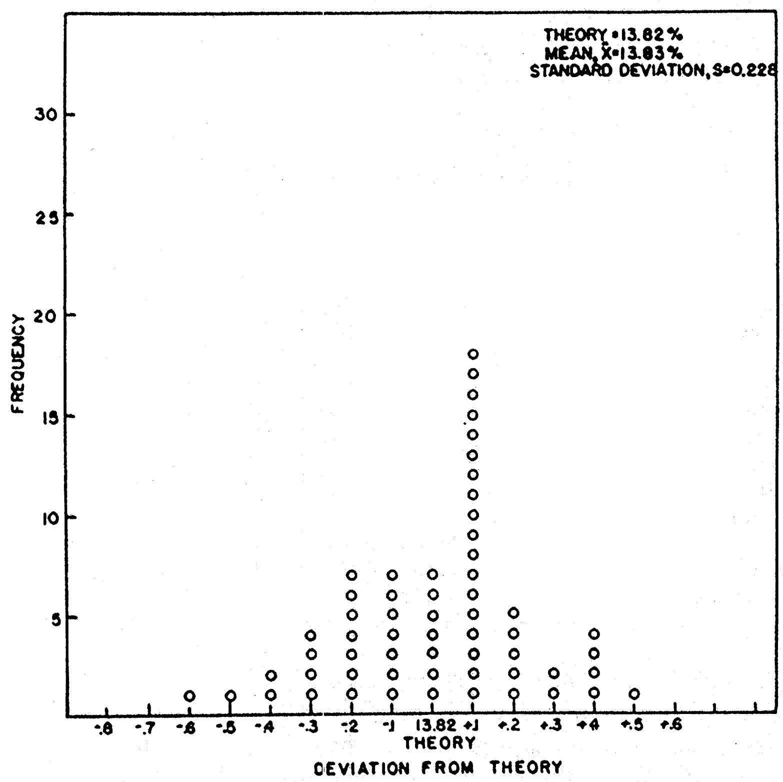


FIG. 14.—Histogram of Dumas nitrogen values for benzyl-iso-thiourea hydrochloride.

below theory, and nicotinic acid has a second mode 0.4% below the theoretical value. The questionnaires indicated that most of the lower values were obtained by methods which used temperatures below 650° C. Consequently, the nitrogen values for nicotinic acid obtained by methods with temperatures above 650°C. were tested and found to be representative of a normal population. Although the data for sample 2 were not as skewed as for sample 1, they were treated in the same manner and with the same result.

The data shown in Table 4 were obtained by methods in which a temperature above 650°C. was used. No comparisons can be made, since the number of representative analyses is too small and there is too much overlapping of values.

The data for sample 1 indicate that the better methods would use, in addition to temperatures above 650°C., a gas sample burner with two burnings, a gasometer, and 1.1% gas volume correction, and would take the temperature of the air as that of the gas in the nitrometer. The data for sample 2 indicate that the methods which use the alternates of these

TABLE 4.—*Nitrogen data obtained for samples 1 and 2 by the Dumas method*

	NICOTINIC ACID (1)			BENZYL-ISO-THIOUREA HYDROCHLORIDE (2)		
	NUMBER OF SAMPLES	$\overline{X}$	S	NUMBER OF SAMPLES	$\overline{X}$	S
		per cent			*per cent*	
Gas burner	18	11.41	0.055	16	13.97	0.152
Electrical burner	20	11.32	0.134	23	13.90	0.190
Sample burned twice	18	11.41	0.055	16	13.97	0.152
Sample burned once	20	11.32	0.134	23	13.90	0.190
Gasometer	17	11.41	0.063	17	13.97	0.145
No gasometer	21	11.33	0.134	22	13.90	0.197
1.1% gas volume correction	15	11.41	0.071	15	13.97	0.152
2% gas volume correction	12	11.29	0.158	12	13.78	0.155
Temperature measured in air	12	11.43	0.067	21	14.03	0.145
Temperature measured in liquid	26	11.33	0.121	18	13.81	0.145
Total samples	38	11.36	0.118	39	13.93	0.179
Theoretical values		11.38			13.82	

operations would be the better. This reversal of suitability of methods is improbable, and can no doubt be accounted for. Of these operations, the only arbitrary means of increasing or decreasing the per cent nitrogen is the correction applied to the gas volume. The mean nitrogen value for sample 2 after a 1.1% volume correction had been made, was too high by 0.15%. It seems unlikely that any of the other variations listed would cause this high value. Therefore, a 2% correction, which would give a mean value only 0.04% low, is apparently the more nearly correct and perhaps should be used in all cases. The reason that the 1.1% correction gave values nearer the theoretical value for nicotinic acid may be that this smaller correction compensated for some nitrogen not recovered from this refractory material.

Summary for Dumas nitrogen.—The study has shown that for satisfactory results by the Dumas method, the minimum temperature is 650°C.

A 2% volume correction appears to be required instead of the often-used 1.1% correction, but further work is necessary to prove this point and to establish preferences for other variations in the procedure.

The values submitted by the various collaborators for carbon, hydrogen, and nitrogen are presented graphically in Figures 15 to 19. No attempt has been made to analyze the values obtained from each laboratory separately.

The collaborators in these studies are listed below.

Collaborators on nitrogen analysis:

Alicino, J. F., Squibb Institute for Medical Research
Jones, G. A., E. I. du Pont de Nemours and Company
Brunner, A. H., Ansco
Ketchum, D. E., Eastman Kodak Company
Powers, D. A., Celanese Corporation of America
Ogg, C. L., Eastern Regional Research Laboratory
Hegeman, B., The Texas Company
Dutton, C. D., Picatinny Arsenal
Grodsky, J., Ortho Research Foundation
Milner, R. T., Northern Regional Research Laboratory
Blackman, S. W., The Wellcome Research Laboratories
Sundberg, O. E., Calco Chemical Division, American Cyanamid Company
Wagner, E. C., University of Pennsylvania

Collaborators on carbon and hydrogen analysis:

Kuck, J. A., American Cyanamid Company
Owens, J. K., E. I. du Pont de Nemours and Company
Sievers, D. C., Tennessee Eastman Corporation
Rachele, J. R., Cornell University Medical College
Paulson, R. A., National Bureau of Standards
Brown, L. E., Southern Regional Research Laboratory
Huffman, E. W. D., Huffman Microanalytical Laboratories
Conard, V. A., Oakwold Laboratories
Feldman, J. R., General Foods Corporation
Hallett, L. T., General Analine and Film Corporation
Hynes, W. A., Fordham University
Clark, H. S., Illinois State Geological Survey
Aluise, V. A., Hercules Powder Company
Butler, A. Q., Mallinckrodt Chemical Works
Shreve, L. S., Smith, Kline and French Laboratories
Streeter, K. B., Sharp and Dohme, Inc.
Steyermark, Al, Hoffman-La Roche, Inc.
Means, J. A., Charles Pfizer and Company, Inc.

RECOMMENDATIONS*

It is recommended that studies be continued on methods for the micro determination of carbon and hydrogen, and for nitrogen, by the Kjeldahl and Dumas procedures. Proposals based on the results of this year's work will be made concerning the procedures to be studied in the next year.

* For report of Subcommittee C and action of the Association, see *This Journal*, 32, 56 (1949).

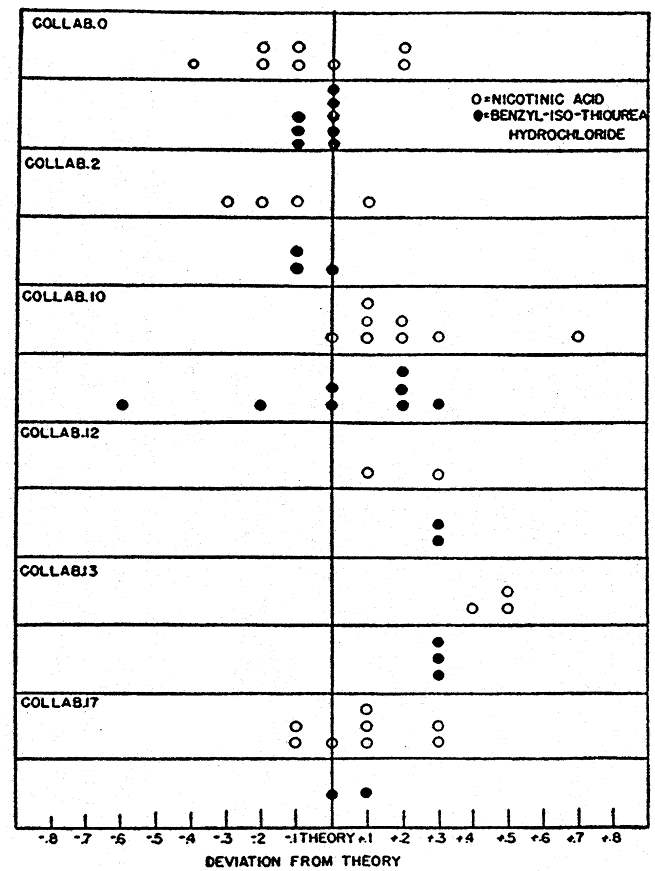


FIG. 15.—Distribution of carbon values from the different collaborators.

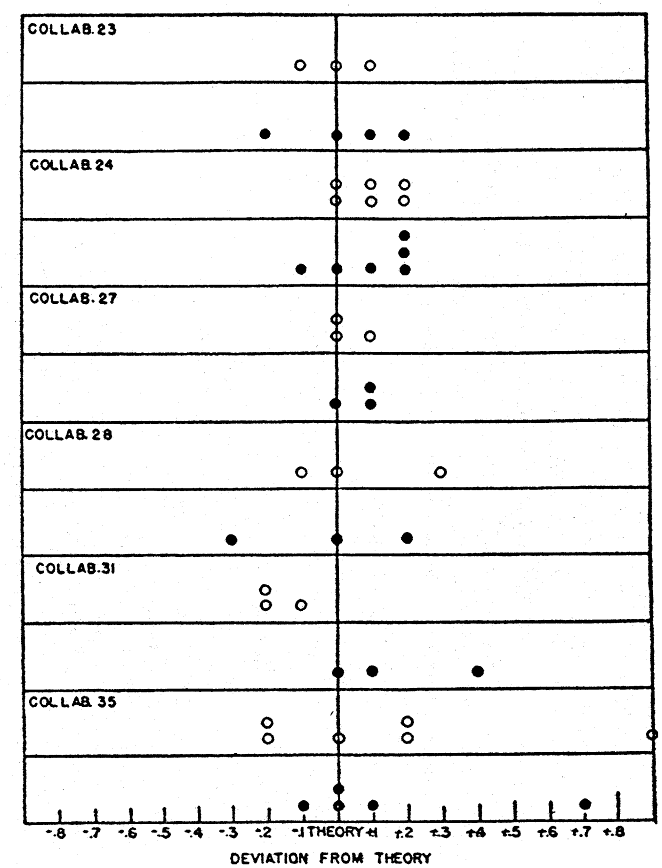


FIG. 15.—Continued.

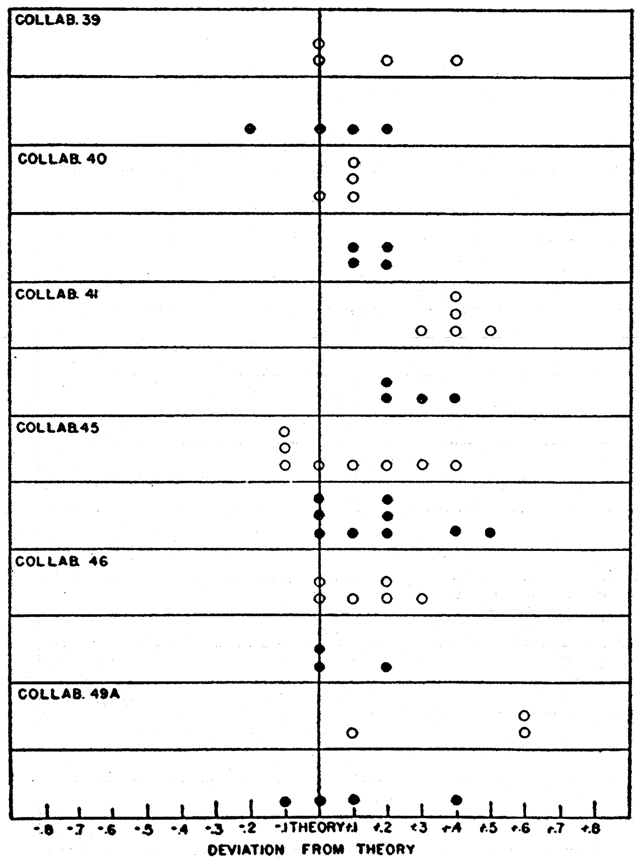
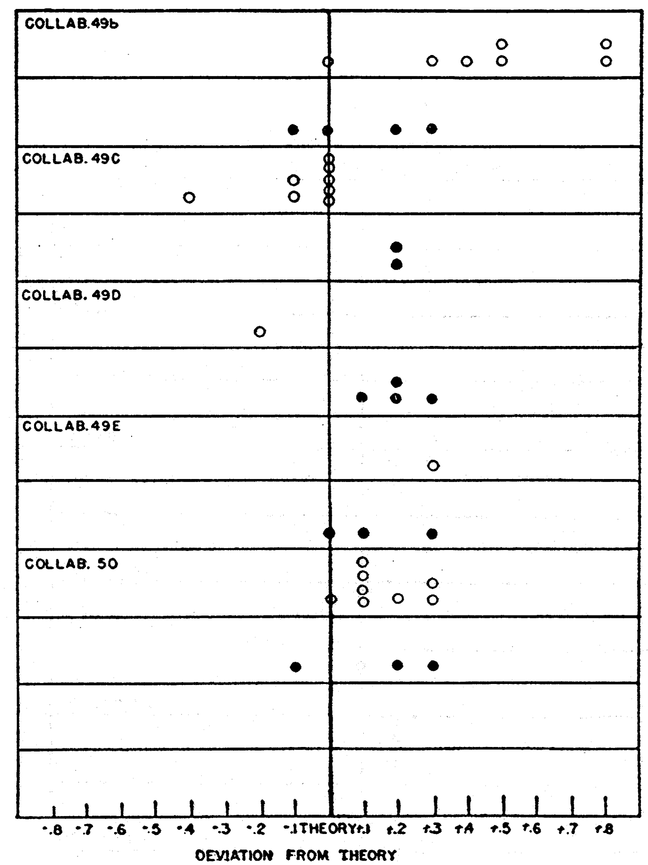


Fig. 15.—Continued.



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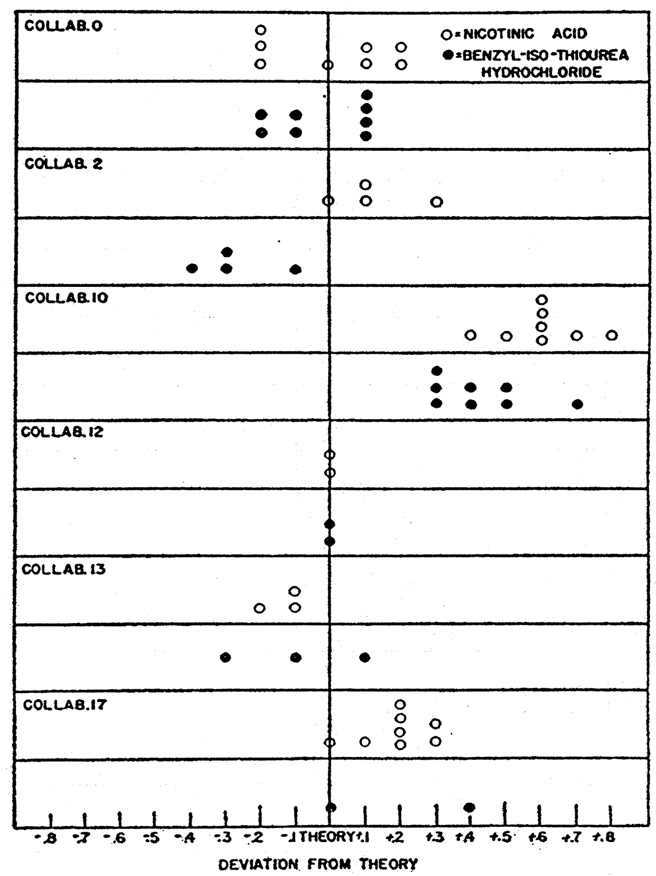

FIG. 16.—Distribution of hydrogen values from the different collaborators.

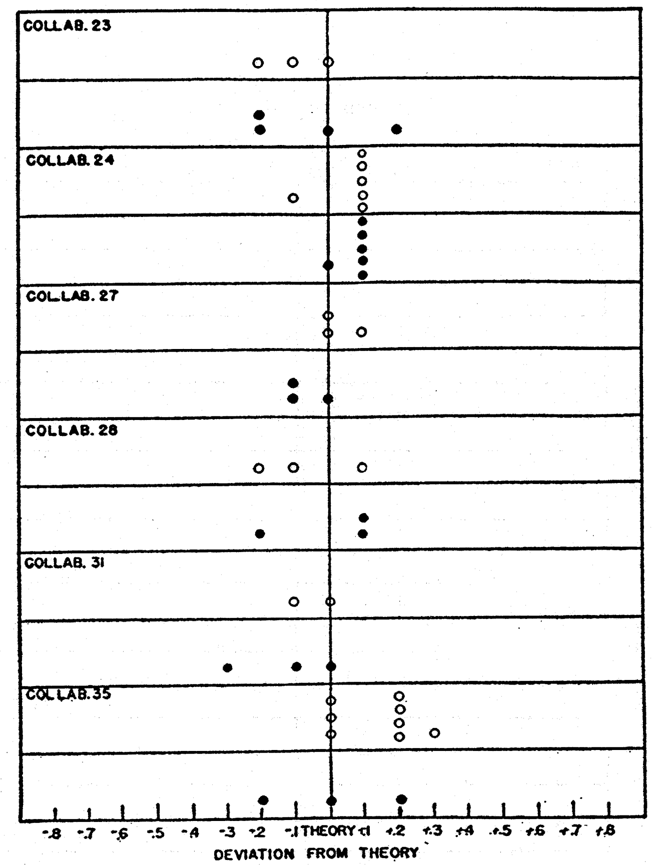

FIG. 16.—Continued.

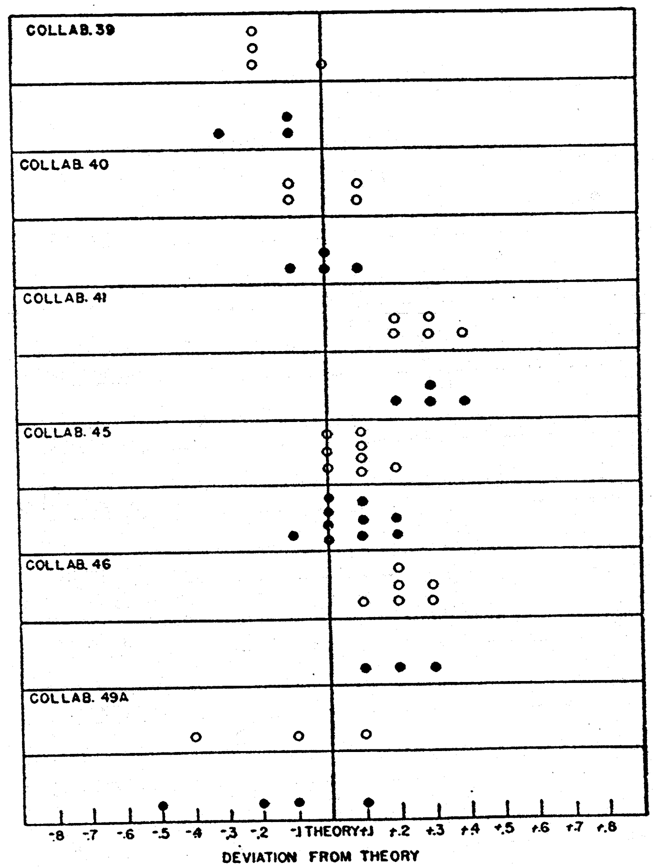

Fig. 16.—Continued.

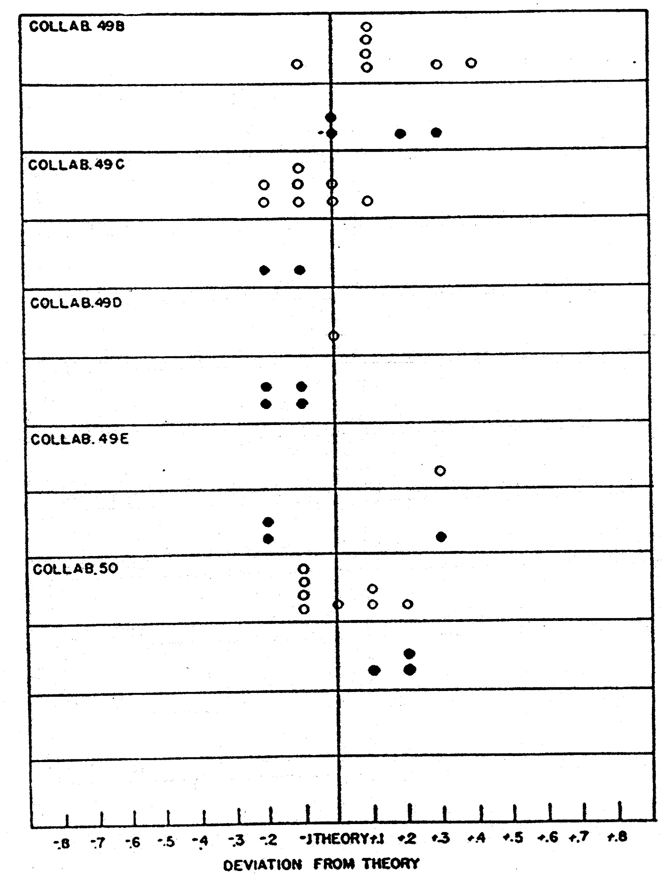

Fig. 16.—Continued.

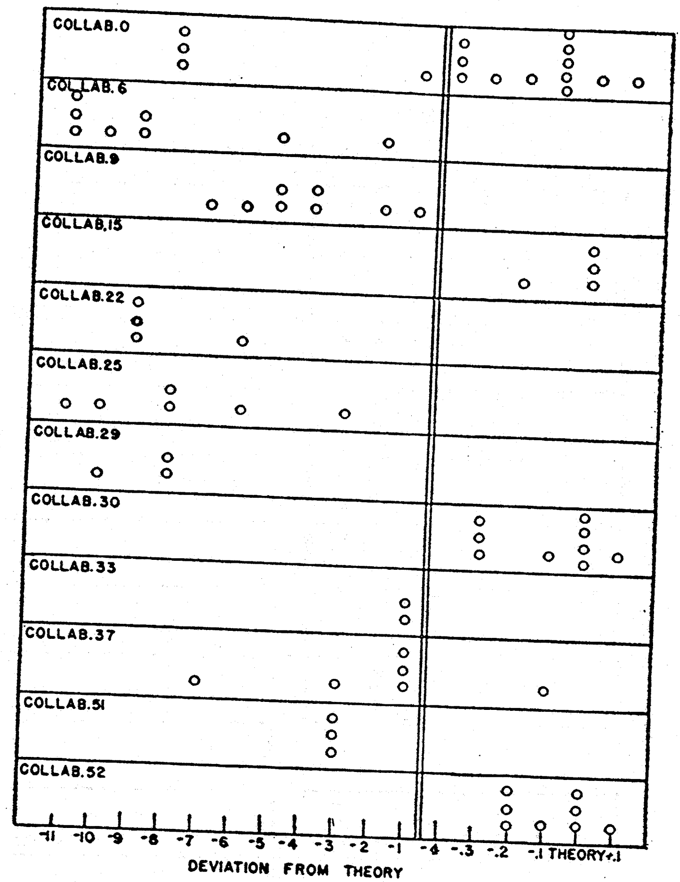

FIG. 17.—Distribution of Kjeldahl nitrogen values for nicotinic acid from the different collaborators.

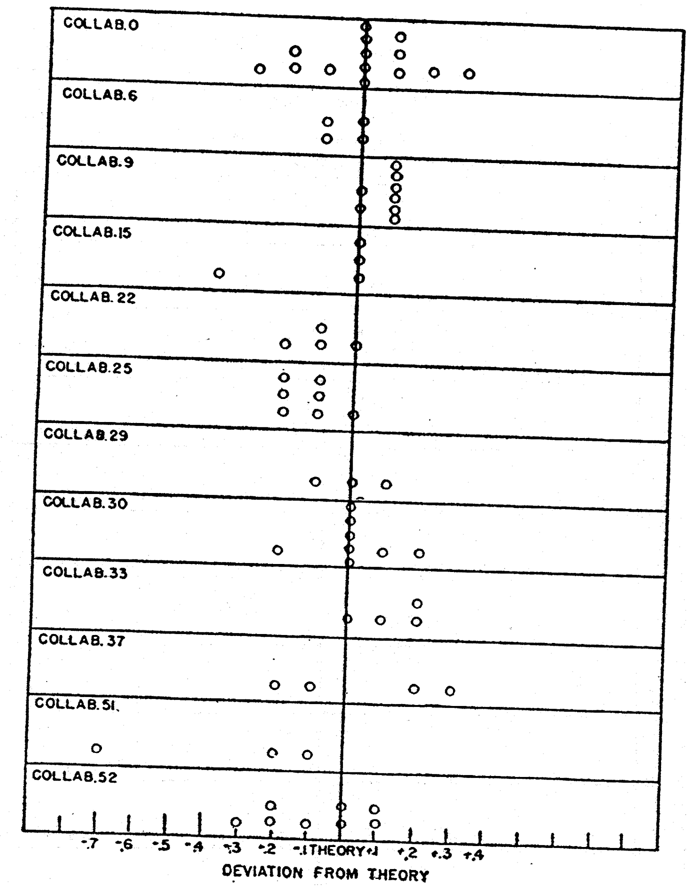

FIG. 18.—Distribution of Kjeldahl nitrogen values for benzyl-iso-thiourea hydrochloride from the different collaborators.

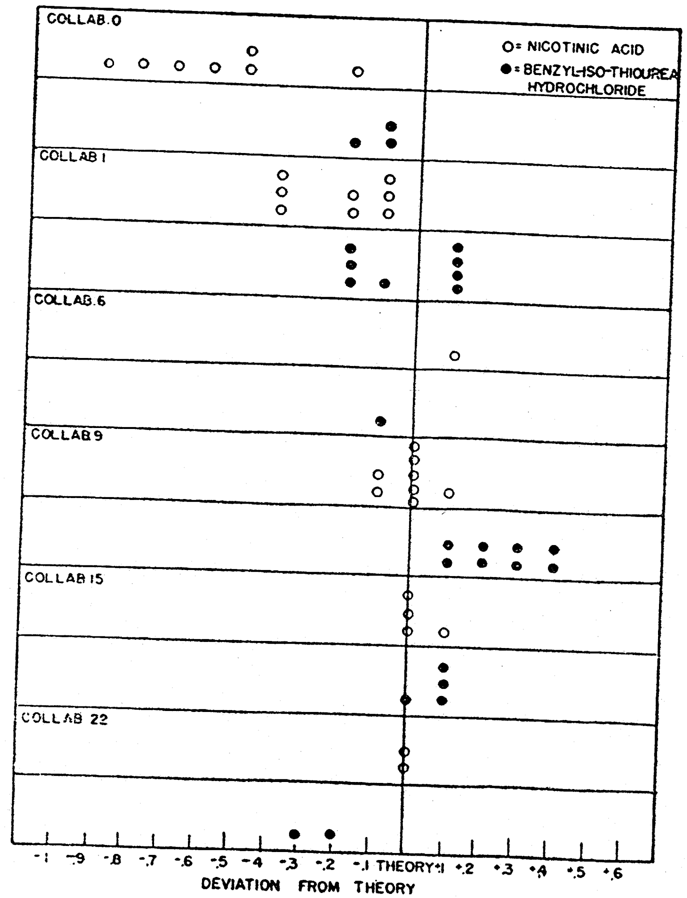

FIG. 19.—Distribution of Dumas nitrogen values from the different collaborators.

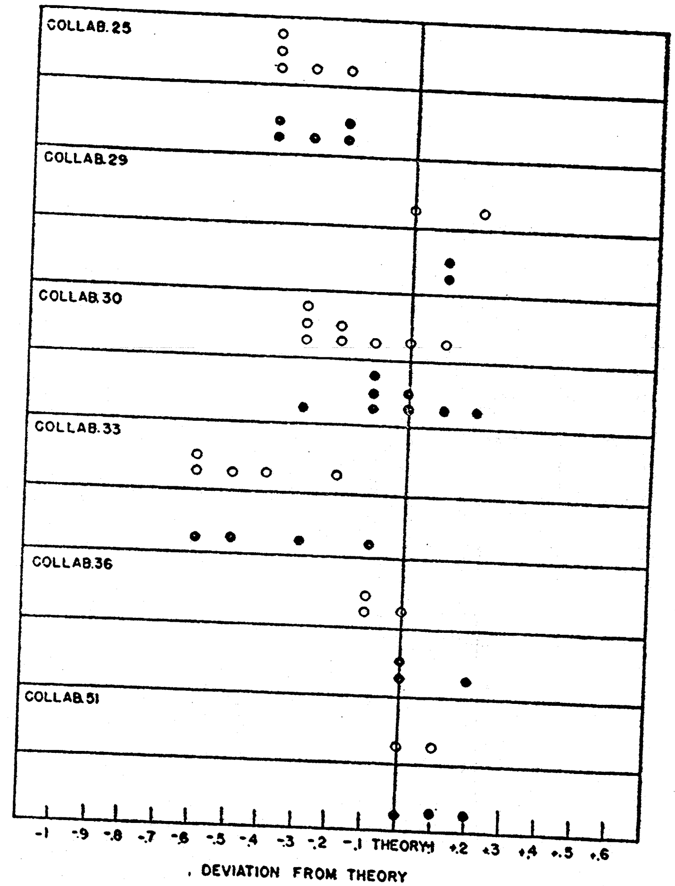

FIG. 19.—Continued.

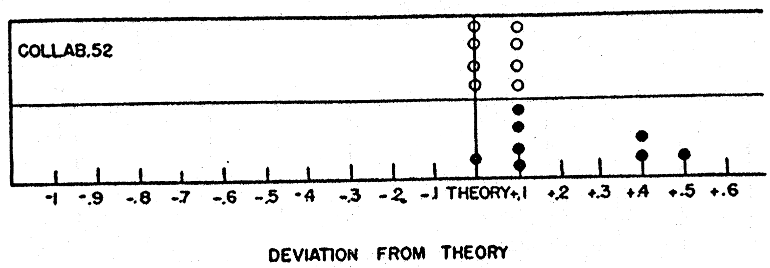

FIG. 19.—Continued.